reactants (mole ratio of silane agent to Bis-GMA) and the reaction conditions; and 4) formulation of the composites without the need for presilanization of the filler phase.

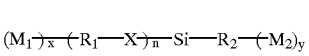
One study, Antonucci et al., *J. Dent. Res.* 65, 219, Abstract 451 (1986) describes the synthesis of such silylated resins. Thus, the reaction of Bis-GMA with monofunctional silanes such as 3-methacryloxypropyldimethyl chloro- or ethoxy-silanes under base catalysis yields the corresponding disilyl ether of Bis-GMA. The resultant tetramethacrylate exhibits a much lower viscosity than Bis-GMA despite its greater bulk (molecular weight of 880). It is expected from structure-property considerations that these silyl derivatives, and especially the ones described herein, will exhibit enhanced toughness, fatigue resistance, and hydrophobicity that will result in polymeric materials of improved durability and environmental resistance.

Hydrophobic dimethacrylate monomers derived from bisphenol A and analogs thereof have also been described in U.S. Pat. No. 3,860,556 to Taylor as a means of improving the dimensional stability of dental composites in the aqueous oral environment. The trimethylsilyl reagents employed in the derivatization are monofunctional, however, in the sense that they can react only once with a hydroxyl group.

SUMMARY OF THE INVENTION

It is an object of the present invention to provide silylated resins having potential for use as adhesive binders for composites, and in sealant and adhesive dental applications. The present invention advantageously employs multifunctional silanes such as alkyl or aryl trifunctional silanes, with good leaving groups attached to the silicon, such as OCH_3, OCH_2CH_3, O_2CCH_3, and $N(CH_3)_2$ and more specifically for example, trialkoxyorganosilanes such as trimethoxyorganosilanes, which can react once, twice, or three times with hydroxyl, amino, and carboxyl groups, as well as other protic functional groups, and thereby, yield a variety of resins.

Accordingly, the invention relates to a silylated resin represented by the general formula (1):

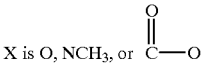


(I)

in which:

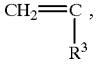
R_1 is an aliphatic, cycloaliphatic, aryl, hydrocarbon, or fluorocarbon group;

R_2 is the same as R or a different aliphatic, cycloaliphatic, aryl, hydrocarbon, or fluorocarbon group;



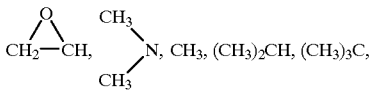
X is O, NCH_3, or

M_1 is



where R_3 is H, CH_3, or CH_2CH_3;

M_2 is the same as M_1 or a different functional or non-functional group selected from the group consisting of:



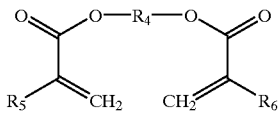
CF_3, and C_6H_5;

n is 1–3;

x is 1–20; and

y is 1–20;

which comprises the reaction product of the exchange reaction of a hydroxylated, aminated, or carboxylated acrylic resin represented by the general formula (II):



(II)

in which:

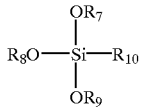
R_4 is an aliphatic, cycloaliphatic, aryl, hydrocarbon, or fluorocarbon group with one or more protic functional groups selected from the group consisting of:

OH, N—H, and CO_2H

R_5 is H or CH_3; and

R_6 is H or CH_3;

with a trialkoxyorganosilane or triacyloxyorganosilane represented by the general formula (III):

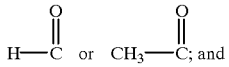


(III)

in which:

R_7, R_8, and R_9 each is:

CH_3, CH_3CH_2, $CH_3CH_2CH_2$, $(CH_3)_2CH$,



or ; and

R_{10} is an aliphatic, cycloaliphatic, or aryl group which can optionally be substituted with a group from the group consisting of an acrylic group, a methacrylic group, an epoxy group, and a substituted amino, hydroxyl, or carboxylic acid group such as an ester or an amide.

The present invention permits the facile synthesis of a wide variety of readily polymerizable silylated resins with a wide spectrum of properties. Because of the presence in many of these resins of pendant, readily-converted silyl functional groups, such as, silyl ether or silyl ester groups of the type shown below: